

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: Roger R. Lesieur et al

Docket No.: C-2407 Cont.

Serial No.: Continuation of 09/512,035

Group: 1764

Filed: Concurrently herewith

Examiner: N. Priesch

For: "Method for Desulfurizing Gasoline or Diesel Fuel for Use in an Internal
Combustion Engine"

PRELIMINARY AMENDMENT

Hon. Commissioner of Patents and Trademarks
Washington, D.C. 20231

Dear Sir:

Please amend the above-identified application as follows.

IN THE SPECIFICATION:

On page 1, before "Technical Field", please insert --This is a continuation of co-
pending USSN 09/512,035, filed December 24, 2000.--.

On page 3 please rewrite the third full paragraph as follows:

"We have discovered that the presence of oxygenates in the gasoline, like MTBE (methyl-tertiary-butyl ether, i.e., $(CH_3)_3COCH_3$), ethanol, or water vapor for example, will prevent rapid deactivation of the nickel catalytic adsorption of organic sulfur compounds from the fuel stream. Ethanol could be an appropriate solution to this problem since it is non-toxic, is not a carcinogen, and is relatively inexpensive and readily available in large supplies as a byproduct of the agriculture industry. Methanol, which would also extend the desulfurizer bed life, is not preferred since it is toxic; while MTBE is likewise not preferred since it is thought to be a carcinogenic compound, and may be banned in certain areas of the United States in the near future by new environmental regulations. Preferred oxygenates are non-toxic and non-carcinogenic oxygen donor compounds, such as ethanol, water vapor or the like. When water is an oxygenate included in the gasoline or diesel fuel mixture being

desulfurized, the water content of the fuel mixture should be in the range of about 3% to about 5% by weight of the fuel mixture."

IN THE CLAIMS:

Please cancel Claims 13-18 without prejudice.

Please rewrite Claims 1, 6 and 9-12 as follows.

1.(amended) A method for desulfurizing a hydrocarbon fuel stream so as to convert the hydrocarbon fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

- a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;
- b) introducing a hydrocarbon fuel stream which contains an oxygenate into said nickel reactant desulfurization station; and
- c) said oxygenate being present in said fuel stream in an amount which is effective to provide an effluent fuel stream at an exit end of said nickel reactant station which effluent fuel stream contains no more than about 0.05 ppm by weight sulfur.

6.(amended) A method for desulfurizing a gasoline or diesel fuel stream so as to convert the gasoline fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

- a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;
- b) introducing a gasoline or diesel fuel stream which contains an oxygenate into said nickel reactant desulfurization station; and
- c) said oxygenate being present in said fuel stream in an amount which is effective to provide an effluent fuel stream at an exit end of said nickel reactant station which

effluent fuel stream contains no more than about 0.05 ppm by weight sulfur.

9.(amended) A method for desulfurizing a gasoline or diesel fuel stream so as to convert the fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

- a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;
- b) introducing a gasoline or diesel fuel stream which contains an oxygenate into said nickel reactant desulfurization station; and
- c) said oxygenate being present in said fuel stream in an amount which is effective to provide a continuous fuel stream at an exit end of said nickel reactant station which continuous fuel stream contains on average no more than about 0.05 ppm by weight sulfur.

10.(amended) A method for desulfurizing a gasoline or diesel fuel stream so as to convert the fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

- a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;
- b) introducing a gasoline or diesel fuel stream which contains an oxygenate into said nickel reactant desulfurization station; and
- c) said oxygenate being converted to isobutylene and methanol by said nickel catalyst in amounts which are effective to inhibit carbon deposition in said nickel catalyst station and provide a continuous fuel stream at an exit end of said nickel reactant station which continuous fuel stream contains no more than about 0.05 ppm by weight sulfur.

11.(amended) A method for desulfurizing a gasoline or diesel fuel stream so as to convert the fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

- 10075670 - 021902
- a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;
 - b) introducing a gasoline or diesel fuel stream which contains an oxygenate into said nickel reactant desulfurization station, said oxygenate being present in said fuel stream in an amount which is effective to provide a low sulfur content fuel stream at an exit end of said nickel catalyst station which low sulfur content fuel stream contains no more than about 0.05 ppm by weight sulfur; and
 - c) said oxygenate being converted to isobutylene and methanol by said nickel reactant during said desulfurizing step, said low sulfur content fuel stream being formed so long as said nickel reactant continues to convert the oxygenate.

12.(amended) A method for desulfurizing a liquid gasoline or diesel fuel stream so as to convert the fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

- a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;
- b) maintaining said nickel reactant desulfurization station at a temperature in the range of about 300°F to about 450°F;
- c) introducing a liquid gasoline or diesel fuel stream which contains an oxygenate into said nickel reactant desulfurization station, said oxygenate being present in said fuel stream in an amount which is effective to provide a low sulfur content fuel stream at an exit end of said nickel reactant station which low sulfur content fuel stream contains no more than about 0.05 ppm by weight sulfur; and
- d) said oxygenate being converted to isobutylene and methanol by said nickel reactant during said desulfurizing step, said low sulfur content fuel stream being formed so long as said nickel reactant continues to convert the oxygenate.

REMARKS

This is a continuation application of co-pending USSN 09/512,035, filed December 24, 2000. Prosecution of the parent application is ongoing. Enclosed are copies of

the parent application as originally filed, and a copy of the declaration for the parent application as originally filed. This amendment includes all of the changes to the claims and to the specification of the parent application which have been made up to the date of filing of this application. A clean copy of the claims as presently amended is enclosed, and a clean copy of the amended paragraph of the specification is also enclosed.

Claims 1-12 are pending in this continuation application. These claims have been rejected in a final rejection in the parent application dated September 24, 2001.

In the final rejection, Claims 1-9 were rejected as being obvious over Setzer et al '746; and Claims 10-12 were rejected as being obvious over Setzer et al '746 in view of Alexander et al. Both of the rejections thus rely on the Setzer et al reference.

In Setzer et al, the desulfurization process is carried out at temperatures in the range of about 500° to 900°F (Col. 3, line 2); and the desulfurized fuel must have a sulfur content of less than about 20 ppm (Col. 3, line 16), since a sulfur content in the desulfurizer bed which above that level indicates that the bed is not functioning properly. .

As noted on page 3 of the office action, there are "Several differences" between the applicants' claimed invention and the contents of the Setzer et al reference.

With respect to Claims 1-9, since the operating temperature range is not specified, there is actually only one difference and that difference is the allowable sulfur content of the desulfurizing bed effluent, which in Setzer et al is anything under 20 ppm and in the instant application is less than 0.05 ppm. Thus the arguments provided by the Examiner regarding the obviousness of the 300°F to 450°F temperature operating range are really irrelevant with respect to the subject matter of Claims 1-9.

Claims 1-9 of the instant application require that the effluent fuel stream exiting the desulfurization bed contain no more than about 0.05 ppm sulfur. The Examiner has

focused on the use of the term "trace" in the Setzer et al patent, and has alleged that "It would have been obvious to one of ordinary skill in the art at the time the invention was made to desulfurize the Setzer et al +++ fuel to any degree required to produce a suitable fuel cell feed, including applicant's' less than 0.05 ppm sulfur +++". (emphasis added). In Setzer et al, it is clearly suggested that a "trace" amount of sulfur is less than 20 ppm. Thus, 19 ppm is a "trace" amount of sulfur as far as Setzer et al is concerned. The Examiner has suggested that one skilled in the art would be able to use the teachings of Setzer et al to achieve an effluent having less than 0.05 ppm sulfur, or in fact any "desired level" which is less than 20 ppm. Thus, following the Examiner's line of reasoning, one skilled in the art could use the teachings of Setzer et al to achieve an effluent having, say, 0.000001 ppm sulfur, since this amount is less than 20 ppm. While making this bold allegation, the Examiner has not pointed out which parameters in Setzer et al would have to be manipulated in order to achieve an effluent having less than 0.05 ppm sulfur. The Examiner is reminded that the rejection of Claims 1-9 is based solely on the contents of the Setzer et al reference, and on nothing else other than Examiner speculation. In order to discharge her burden of proof, the Examiner must present a cogent analysis of the Setzer et al reference pointing out what parameters would be varied by one skilled in the art to achieve the claimed less than 0.05 ppm sulfur content effluent. For example, would one increase (or decrease) the amount of water suggested by Setzer et al; or would one increase (or decrease) the operating temperatures of the Setzer et al desulfurizer? The Examiner must also point out where in the Setzer et al reference she finds these clues as to how to achieve the claimed less than 0.05 ppm sulfur levels.

Claims 10-12 require the formation of methanol and isobutylene during the desulfurization process, and Claim 12 requires that the desulfurizer station be maintained at a temperature in the range of about 300°F to about 450°F. The Examiner alleges that a temperature range of 500°F-900°F (Setzer et al) is close enough to a temperature range of 300°F to 450°F (this application) to convey to one of ordinary skill in the art an expectation that the Setzer et al process would work at the lower temperatures claimed in the instant application.

10076670-021902

The 500°F to 900°F operating range is the only operating range disclosed in Setzer et al '746, and we submit that 500°F is not close enough to 450°F to suggest that the Setzer et al desulfurizer could be operated at the lower temperature. In Setzer et al '746 "breakthrough" is a condition which is not desirable. It, in fact, denotes failure of the sulfur removal bed, as far as Setzer et al '746 is concerned. It is thus elementary that postponement of breakthrough is most desirable. FIG. 2 in Setzer et al '746 provides clues as to the relationship between the time to reach breakthrough (in hours) and the operating temperature (in degrees F) of the desulfurizing bed. FIG. 2 indicates that breakthrough will occur after about 9 hours from start up at an operating temperature of 500°F, and the longest breakthrough time (about 16 hours) will occur at an operating temperature of about 600°F. FIG. 2 also indicates that the breakthrough time decays at temperatures between 600°F and 900°F. FIG. 2 indicates that breakthrough times at 900°F and 500°F are about the same, i.e., about 9 to 10 hours. The Examiner has alleged that the claimed temperature range of 300°F to 450°F is "close enough" to the prior art temperature range of 500°F to 900°F so that one of skill in the art would have expected "the same properties". The Examiner in essence is ignoring the specific working temperature range of 500°F to 900°F of Setzer et al and is characterizing the reference as disclosing an elevated temperature which in her view encompasses any temperature which would accomplish an effective desulfurization. What temperature, in the Examiner's view is not "elevated".

The Examiner has not bothered to define what she means by "elevated", thus we are left in the dark as to what she is talking about. In the rejection, the Examiner implies that Applicants must demonstrate that the 450°F upper limit of the claimed temperature range shows something "unexpected". There is no requirement for anything "unexpected" in Title 35 of the patent statute. Kansas Jack, Inc. v Kuhn et al 219 USPQ 857 (CAFC 1983). With all due respect, the Examiner would be well advised to forget about "unexpected results" when formulating §103 rejections. A §103 rejection requires that the modification be obvious in view of the prior art. A modification is "obvious" if the prior art provides some motivation to incorporate the modification, or some indication that the modification would be desirable. The Examiner has the

burden of proof to point out where that motivation or desirability is provided in the prior art. In Ex parte Levy, 17 USPQ2d 1461 (US PTO Bd. Pat. App. & Int. 1990), the Board held that the initial burden of establishing a *prima facie* basis to deny patentability rests upon the examiner. See also In re Carleton, 202 USPQ 165 (CCPA 1979); and In re Piasecki, 223 USPQ 785 (Fed. Cir. 1984). This burden can only be discharged by establishing a factual basis for the finding of non-patentability. Examiner's conjecture or conclusionary assertions do not provide the necessary factual basis.

The Examiner has necessarily alleged that the upper end of the claimed temperature range (450°F) is close enough to the lower end of the prior art temperature range (500°F) so that one of ordinary skill in the art would be motivated by the prior art, or would believe that the claimed 450°F temperature would be desirable to use in the performance of the prior art Setzer et al procedure. We interpret the Examiner's position as alleging that the use of a 450°F operating temperature in the Setzer et al '745 procedure would be desirable and should be used. Looking at FIG. 2 of Setzer et al '745 we note that the graph indicates that there is a linear decrement in the breakthrough time between the operating temperatures of 600°F and 900°F, and that the use of temperatures which are greater than 600°F are more desirable than the temperature of 500°F. Operating temperatures below 500°F, if extrapolated from FIG. 2, would provide a breakthrough time which appears to decline logarithmically, and which appears to be at zero hours before reaching an operating temperature of 450°F. Thus the only correlation between operating temperatures and the achievement of the undesirable breakthrough shown in Setzer et al '745 is shown in FIG. 2 and indicates that it would not be desirable to utilize an operating temperature which is below 500°F, and certainly not one which is as low as 450°F. Setzer et al '745 thus does not suggest that an operating temperature of less than 500°F would be desirable, or even useful.

Thus the teachings of Setzer et al '745 would not "motivate" one skilled in the art to utilize an operating temperature which is included in the range of temperatures that

are recited in Claim 12 of the instant application, irregardless of how "close" the upper end of the range of the claimed operating temperatures is to the lower end of the range of operating temperatures set forth in the prior art.

Stated more succinctly, in the instant case, "close enough" is not good enough. If the Examiner can present a well reasoned argument which proves that, in her words, "close enough" is good enough, then we would like to consider it.

B) Claims 10-12 stand rejected as being obvious over Setzer et al '746 in view of Alexander et al.

The flaws in the Examiner's analysis of Setzer et al '746 relating to the sulfur content of the desulfurizing bed effluent, and the desulfurizing bed operating temperatures are noted above, and are reiterated in connection with this rejection.

Alexander et al is relied upon by the Examiner for a showing that "conventional gasoline marketed in large metropolitan areas contains oxygenates including methanol, ethanol and MTBE. See column 1, lines 1-25.". What the Examiner is ignoring is the contents of the third full paragraph in Col. 1 (lines 26-33) of Alexander et al wherein the patent notes that there are many problems associated with the use of methanol and ethanol as a gasoline additive, and therefore the industry would not suggest the use of either of these two additives in gasoline. This patent thus teaches away from the use of ethanol and/or methanol as an additive in gasoline. Additionally, the use of MTBE as a gasoline additive has recently come under attack since there is evidence that MTBE is a cancer-causing agent. MTBE in gasoline has or will be banned in California, and will be banned in Connecticut and elsewhere, in the near future.

The Examiner must concede that there are gasolines that do not include MTBE, or any other oxygenates, presently in use in the United States. Thus, there are presently gasolines which include MTBE, and gasolines which do not include MTBE or any other oxygenates. This being the case, the Examiner has the burden of proof in

showing that one desiring to desulfurize a gasoline or diesel fuel stock would be motivated to use a fuel stock which includes an oxygenate. The Examiner's rejection can thus be characterized as a "could be" case of obviousness. The mere fact that prior art could be modified does not make the modification obvious unless the prior art suggested the desirability of the modification. See: In re Gordon, 221 USPQ 1125 (CAFC 1984); In re Grabiak, 226 USPQ 870 (CAFC 1985); and In re Sernaker, 217 USPQ 1 (CAFC 1983). Neither Alexander et al nor Setzer et al provides the necessary motivation or suggests the desirability to use a fuel stock that must include an organic oxygenate in the desulfurizing process.

Regarding the specific subject matter of Claims 10-12, these claims all require a desulfurized fuel stream which contains no more than about 0.05 ppm sulfur. Alexander et al does not enable the achievement of such a result. Regarding Claims 10-12, these claims all require that the oxygenate be converted to another compound by the nickel reactant bed. Neither Setzer et al nor Alexander et al suggest the desirability of using a nickel reactant bed to convert an oxygenate to another compound of the types recited in these claims.

The Examiner's reliance in the final rejection on vague phrases such as "trace amounts", "as much as possible", "any temperature", "an effective desulfurization", "elevated temperature", and so forth, as grounds for the rejections does not address the issues here. The claims in this patent application do not recite limitations such as "trace amounts", "as much as possible", "any temperature", "an effective desulfurization" or "elevated temperature".

The final rejection also includes a provisional obviousness-type double patenting rejection which is based on co-pending patent application Serial No. 09/470,483. Applicants would be disposed to file a terminal disclaimer at an appropriate time during the prosecution of this application to deal with this rejection.

Respectfully submitted,



William W. Jones

Attorney for Applicant

Reg. No. 24,607

6 Juniper Lane

Madison, CT 06443

(203) 245-2418

Date 1-16-02

10075570-021902

Clean Copy of Claims as Amended

1. A method for desulfurizing a hydrocarbon fuel stream so as to convert the hydrocarbon fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

- a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;
- b) introducing a hydrocarbon fuel stream which contains an oxygenate into said nickel reactant desulfurization station; and
- c) said oxygenate being present in said fuel stream in an amount which is effective to provide an effluent fuel stream at an exit end of said nickel reactant station which effluent fuel stream contains no more than about 0.05 ppm by weight sulfur.

6. A method for desulfurizing a gasoline or diesel fuel stream so as to convert the gasoline fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

- a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;
- b) introducing a gasoline or diesel fuel stream which contains an oxygenate into said nickel reactant desulfurization station; and
- c) said oxygenate being present in said fuel stream in an amount which is effective to provide an effluent fuel stream at an exit end of said nickel reactant station which effluent fuel stream contains no more than about 0.05 ppm by weight sulfur.

9. A method for desulfurizing a gasoline or diesel fuel stream so as to convert the fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

- a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;
- b) introducing a gasoline or diesel fuel stream which contains an oxygenate into said

10076670-021902

nickel reactant desulfurization station; and

c) said oxygenate being present in said fuel stream in an amount which is effective to provide a continuous fuel stream at an exit end of said nickel reactant station which continuous fuel stream contains on average no more than about 0.05 ppm by weight sulfur.

10. A method for desulfurizing a gasoline or diesel fuel stream so as to convert the fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;

b) introducing a gasoline or diesel fuel stream which contains an oxygenate into said nickel reactant desulfurization station; and

c) said oxygenate being converted to isobutylene and methanol by said nickel catalyst in amounts which are effective to inhibit carbon deposition in said nickel catalyst station and provide a continuous fuel stream at an exit end of said nickel reactant station which continuous fuel stream contains no more than about 0.05 ppm by weight sulfur.

11. A method for desulfurizing a gasoline or diesel fuel stream so as to convert the fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;

b) introducing a gasoline or diesel fuel stream which contains an oxygenate into said nickel reactant desulfurization station, said oxygenate being present in said fuel stream in an amount which is effective to provide a low sulfur content fuel stream at an exit end of said nickel catalyst station which low sulfur content fuel stream contains no more than about 0.05 ppm by weight sulfur; and

c) said oxygenate being converted to isobutylene and methanol by said nickel reactant during said desulfurizing step, said low sulfur content fuel stream being formed so long as said nickel reactant continues to convert the oxygenate.

12. A method for desulfurizing a liquid gasoline or diesel fuel stream so as to convert the fuel stream into a low sulfur content fuel, which low sulfur content fuel is suitable for use in an internal combustion engine, said method comprising the steps of:

- a) providing a nickel reactant desulfurization station which is operative to convert sulfur contained in organic sulfur compounds contained in the fuel stream to nickel sulfide;
- b) maintaining said nickel reactant desulfurization station at a temperature in the range of about 300°F to about 450°F;
- c) introducing a liquid gasoline or diesel fuel stream which contains an oxygenate into said nickel reactant desulfurization station, said oxygenate being present in said fuel stream in an amount which is effective to provide a low sulfur content fuel stream at an exit end of said nickel reactant station which low sulfur content fuel stream contains no more than about 0.05 ppm by weight sulfur; and
- d) said oxygenate being converted to isobutylene and methanol by said nickel reactant during said desulfurizing step, said low sulfur content fuel stream being formed so long as said nickel reactant continues to convert the oxygenate.

10075670-021002

Clean Copy of Specification Paragraphs as Amended

On page 3, the third full paragraph:

We have discovered that the presence of oxygenates in the gasoline, like MTBE (methyl-tertiary-butyl ether, i.e., $(\text{CH}_3)_3\text{COCH}_3$), ethanol, or water vapor for example, will prevent rapid deactivation of the nickel catalytic adsorption of organic sulfur compounds from the fuel stream. Ethanol could be an appropriate solution to this problem since it is non-toxic, is not a carcinogen, and is relatively inexpensive and readily available in large supplies as a byproduct of the agriculture industry. Methanol, which would also extend the desulfurizer bed life, is not preferred since it is toxic; while MTBE is likewise not preferred since it is thought to be a carcinogenic compound, and may be banned in certain areas of the United States in the near future by new environmental regulations. Preferred oxygenates are non-toxic and non-carcinogenic oxygen donor compounds, such as ethanol, water vapor or the like. When water is an oxygenate included in the gasoline or diesel fuel mixture being desulfurized, the water content of the fuel mixture should be in the range of about 3% to about 5% by weight of the fuel mixture.